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TWO-PROTON RADIOACTIVITY

(Prospects of Detection and Study)

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1. INTRODUCTION

SPONTANEOUS transformations of chemical elements have been studied in tens and even hundreds of examples for each of three types of disintegration: α decay, β decay (β^- decay, β^+ decay, and orbital electron capture), and spontaneous fission. Rutherford also suggested the possibility of spontaneous transformations involving proton emission. B. S. Dzhelepov made the first detailed analysis of the problem of proton radioactivity and considered the prospect of its detection in 1951.[1] However, even proton radioactivity does not exhaust all the conceivable forms of spontaneous transformations of the elements. Proton pairing in nuclei leads to a fifth possible type of transformation—two-proton *radioactivity*, which will be discussed in the present article.

Let $B_{odd\text{-}p}$ be the attachment energy of an additional odd proton to a nucleus $_{z=2m}M_N^A$ (in other words, the proton binding energy in a nucleus $_{z=2m+1}M_N^{A+1}$). Because of the energy gained through proton pairing the attachment energy of an additional even proton $B_{even\text{-}p}$ (i.e., the proton binding energy in a nucleus $_{z=2m+2}M_N^{A+2}$) usually exceeds $B_{odd\text{-}p}$ despite the opposing growth of Coulomb repulsion energy. Neglecting this change of the Coulomb energy, we shall define the proton pairing energy E_{pair} as the difference

$$B_{even\text{-}p\,(2m+2)} - B_{odd\text{-}p\,(2m+1)} = E_{pair} > 0$$

(ordinarily $E_{pair} \approx 2$ MeV).

The binding energy of two protons in $_{z=2m+2}M_N^{A+2}$ is obviously $B_{2p} = B_{even\text{-}p} + B_{odd\text{-}p} = 2B_{even\text{-}p} - E_{pair} = 2B_{odd\text{-}p} + E_{pair}$. Therefore, when $B_{odd\text{-}p} < 0$, i.e., $B_{even\text{-}p} < E_{pair}$, a pair of protons is more easily detached from an even-Z nucleus than only a single "even" proton. If $B_{even\text{-}p} < 1/2E_{pair}$ (in other words, the decay energy of an odd-Z nucleus $Q_{odd\text{-}p} = -B_{odd\text{-}p} > 1/2E_{pair}$), an even-Z nucleus becomes energetically unstable with respect to the simultaneous emission of two protons. The two-proton decay energy is $Q_{2p} = -B_{2p} > 0$, whereas the binding energy of a single "even" proton can still remain positive: $0 < B_{even\text{-}p} < 1/2E_{pair}$.

In 1957 it was demonstrated for the first time that a certain nucleus does not exist because of two-proton instability;[2] other authors continued the study.[3] The nucleus in question is Be^6, whose decay scheme based on the data compilation in[4] is shown in Fig. 1.

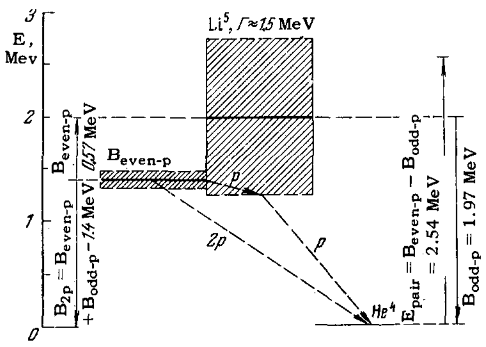

FIG. 1. Decay scheme of Be^6 ($\Gamma \lesssim 150$ keV).

Here $B_{even\text{-}p}$ (Be^6) = 0.57 MeV and $Q_{odd\text{-}p}$ (Li^5) = $-B_{odd\text{-}p}$ = 1.97 MeV, i.e., Q_{2p} (Be^6) = 1.4 MeV and E_{pair} = 2.54 MeV. The Be^6 lifetime against the decay $Be^6 \rightarrow He^4 + 2p$ is $\tau \gtrsim 4 \times 10^{-21}$ sec; this is almost two orders of magnitude greater than the characteristic nuclear time ($\sim 10^{-22}$ sec), which is of the order of the nucleonic period of revolution within a nucleus and is about 10 times greater than the mean lifetime of Li^6 ($\tau \approx 4 \times 10^{-22}$ sec), which decays into a proton and α particle. Be^6 is apparently not the only example of a nonexisting nucleus because of 2p instability (the "instantaneous" breakdown of a two-proton-unstable system). Thus, after analyzing the existence limits of neutron-deficient isotopes of light elements, Ya. B. Zel'dovich[5] expressed doubts regarding the existence of three more even-Z nuclei—O^{12}, Ne^{16}, and Mg^{19}—because of their possible two-proton instability. The experiments of Vlasov et al. and the predictions of Zel'dovich did not touch upon the possibility that two-proton radioactivity can occur and therefore did not consider the properties of this type of radioactive decay. They were concerned only with the question whether a few light nuclei "exist or do not exist," depending upon whether the binding energy of a proton pair in these nuclei is positive or negative. For Be^6 nonexistence was proved experimentally; in the cases of O^{12}, Ne^{16}, and Mg^{19} the possibility of the same answer was suggested, i.e., the possibility that these nuclides are unstable with respect to the simultaneous emission of two protons.

It is clear, however, that energetic instability is only a necessary, but not a sufficient, condition for radioactive decay. This instability is in itself not equivalent to radioactivity.

Many dozens of heavy-element isotopes are energetically unstable with respect to α decay and spontaneous fission, yet are among the stable nuclei. On the other hand, we know many examples (especially among light nuclei) where the lifetime of an energetically unstable system is so short that we are not concerned with the existence of a given radioactive isotope, but rather with the metastable states of a system having a given number of nucleons and a given isotopic spin. These examples include not only Li^5 and Be^6, but also Be^9 (known since 1951[6]), which is unstable with respect to the emission of a strongly sub-barrier proton from the ground state ($\Gamma \leq$ 750 eV[7]) but is not, of course, considered radioactive although its lifetime is almost four orders of magnitude greater than the characteristic nuclear time. The classification of different unstable nuclear states has recently been reviewed in Usp. Fiz. Nauk. by A. I. Baz', Ya. B. Zel'dovich, and the present author;[8] therefore this subject will not be considered here in greater detail. It may be mentioned, however, that in order to avoid confusing spontaneous and induced nuclear transformations, radioactivity, and the decay of compound nuclei in nuclear reactions, we customarily limit the concept of radioactive decay to the experimentally measurable times $\tau > 10^{-12}$—10^{-10} sec. Dzhelepov kept this convention in mind in going from the obvious consequence of neutron deficiency, i.e., proton instability, to a specific analysis of the existence and properties of proton radioactivity.[1]

In the case of two-proton decay the transition from hypotheses concerning nuclear instability to the consideration of possible radioactivity is somewhat more complicated than for the relatively trivial one-proton decay. Two-proton radioactivity is a three-body problem with three two-body Coulomb interactions or, if the p-p Coulomb interaction is neglected, it is a problem concerning the simultaneous passage of two protons through the potential barrier surrounding a nucleus. How is this passage affected by energy exchange between the protons and by pairing or depairing under the potential barrier? How, as a result, is the 2p-decay energy related to the lifetime of two-proton-unstable nuclei? Only by answering these questions can we reach any definite conclusions regarding the possibility that two-proton radioactivity exists, or about such a very important characteristic as the energy correlation of the protons.

These questions were answered in[9,10]. After comparing pairing energies with the rate of diproton tunneling as a whole or that of two independent protons, it was concluded[9] that "for even-Z isotopes instability with respect to the simultaneous emission of two protons can occur even when the binding energy of a single proton is positive, as in Be^6, for example. In the presence of a Coulomb barrier this instability can lead to two-proton radioactivity in isotopes that are stable against both proton and α decay."

In[9,10] and in the review article[11] very simple relations based on isotopic invariance were derived, which enabled very accurate predictions of all basic properties exhibited by neutron-deficient isotopes of light elements. These relations, which we shall consider again in the present review, were used to investigate a few specific examples of possible 2p-radioactive nuclei. Subsequent articles[12-14] contain an elementary theoretical description of the basic properties inherent in the hypothetical new type of radioactive decay when only a Coulomb barrier is present. The possible 2p-radioactive nuclei with $Z < 50$ were listed and the methods of producing them were analyzed. An improved theory of two-proton decay for $l = 0$ based on the theoretical equations for a superfluid nucleus was developed in[15]. In[16] the influence of a centrifugal barrier on 2p decay was examined, while[17] discussed the properties of 2p-radioactive nuclei heavier than tin which are unstable with respect to both one-proton decay and α decay. An experimental search for the new type of radioactivity in Ne^{16} has been reported in[18].

When two-proton radioactivity is compared with other physical phenomena, it is easy to observe a very close analogy with electron tunneling between superconducting and normal metals under the influence of a potential difference exceeding the superconducting gap width.

2. RATE OF TWO-PROTON DECAY

By applying the customary formulas for tunneling through a Coulomb barrier to the diproton as a single particle we arrive at an expression for the exponential term C_{2p} in the 2p-radioactive decay constant $\lambda_{2p} = K_{2p}e^{-2C_{2p}}$ for a nucleus of charge Z and mass A:

$$C_{2p} = \frac{(Z-2)e^2\sqrt{m}}{\hbar}\frac{4}{\sqrt{Q_{2p}}}\left[\arccos x^{1/2} - x^{1/2}(1-x)^{1/2}\right], \quad (1)$$

where m is the proton mass, and

$$x = \frac{Q_{2p}}{U_{\text{Coul }2p}}, \quad U_{\text{Coul }2p} \approx \frac{2(Z-2)}{1.25+(A-2)^{1/3}}\ \text{MeV} \quad (1a)$$

(Q_{2p} will be denoted henceforth simply by Q).

We now ask how this constant is changed for the simultaneous independent emission of two protons. We can reasonably assume that in this case $\lambda_{p_1p_2} \approx K_pe^{-2C_{p_1}}e^{-2C_{p_2}}$, with $K_p \approx 10^{22}$ sec^{-1}, and

$$C_p = \frac{(Z-1)e^2\sqrt{m}}{\hbar}\frac{\sqrt{2}}{\sqrt{Q_p}}\left[\arccos y^{1/2} - y^{1/2}(1-y)^{1/2}\right],$$
$$y = \frac{Q_p}{U_{\text{Coul }p}}, \quad U_{\text{Coul }p} \approx \frac{Z-1}{1+(A-1)^{1/3}}\ \text{MeV} \quad (2)$$

(the subscript p will be dropped henceforth whenever it would appear together with the designations "even," "odd," or the numerals 1 or 2).

We shall neglect the nuclear recoil energy (thus taking the decay energy Q as equal to the energy E of the emitted protons), the proton mass and charge compared with the residual nucleus, and the difference between the proton and diproton sizes in the expression for U_{Coul}.

Let the proton energies be $E_1 = Q_1 = Q(1+\kappa)/2$ and $E_2 = Q_2 = Q(1-\kappa)/2$, respectively.* Then $y_1 + y_2 = 2x$, where $y_1 = x(1+\kappa)$, $y_2 = x(1-\kappa)$, and

$$C_1+C_2=\frac{Ze^2\sqrt{m}}{\hbar}\frac{2}{\sqrt{Q}} \times\left\{\frac{\arccos[x(1+\varkappa)]^{1/2}-[x(1+\varkappa)]^{1/2}[1-x(1+\varkappa)]^{1/2}}{(1+\varkappa)^{1/2}} + \frac{\arccos[x(1-\varkappa)]^{1/2}-[x(1-\varkappa)]^{1/2}[1-x(1-\varkappa)]^{1/2}}{(1-\varkappa)^{1/2}}\right\}, \quad (3)$$

and for $x \ll 1$, i.e., in the most interesting cases,

$$C_1+C_2\approx\frac{Ze^2\sqrt{m}}{\hbar}\frac{2}{\sqrt{Q}}\left\{\frac{\frac{\pi}{2}-2[x(1+\varkappa)]^{1/2}}{(1+\varkappa)^{1/2}}+\frac{\frac{\pi}{2}-2[x(1-\varkappa)]^{1/2}}{(1-\varkappa)^{1/2}}\right\}. \quad (4)$$

It is easily proved that the sum $C_1 + C_2$ is minimal, i.e., decay occurs most probably when $x = 0$ with equal energies of the two emitted protons.[9,10] We now have $C_1 = C_2 = C_p = 1/2\, C_{2p}$, so that

$$(C_1+C_2)_{\min}=C_{2p}\approx\frac{Ze^2\sqrt{m}}{\hbar}\frac{4}{\sqrt{Q}}\left[\frac{\pi}{2}-2x^{1/2}\right]. \quad (5)$$

Therefore, except for the factor before the exponential, the emission of a diproton with the energy Q has the same probability as the simultaneous emission of two protons independently with identical energies Q/2. Jänecke[14] has pointed out that when a diproton has left a nucleus the effective decay energy is reduced by the amount $\epsilon_0 \approx 70$ keV, which is the energy of the virtual nucleon-nucleon singlet level. As a result of replacing Q with $Q - \epsilon_0$, in the case of a pure Coulomb barrier the probability that two protons will be emitted independently is higher than for a diproton, i.e., the diproton breaks apart at the inside boundary of the Coulomb barrier. As we shall see subsequently, when the Coulomb barrier is accompanied by a centrifugal barrier, depairing of the emitted protons takes place under the barrier or even at its outside boundary. The rate of two-proton decay is then represented by (1) with $Q - \epsilon_0$ substituted for Q.

The foregoing treatment of the rate of radioactive decay involving the simultaneous emission of two protons is, of course, only a first approximation, which has been found to be quite accurate. Certainly, several particles participate in the process, which presents a problem having a minimum of three interacting bodies.

*In[9-13] we used the notation $E_1 = (Q/2) + (\kappa Q_2)$ and $E_2 = (Q/2) - \kappa Q$. However, the relations $E_1 = Q(1+\kappa)/2$ and $E_2 = Q(1-\kappa)/2$ are more intuitively clear. We must remember that κ is here twice as large as in the earlier articles.

A more consistent theory of two-proton radioactivity in the case of proton emission with zero orbital angular momentum has been given by Galitskiĭ and Chel'tsov in[15]. The problem was based there on its formal similarity to nucleon pairing in a spherically symmetric nucleus, for which a solution has been obtained several times in connection with the superfluidity of nuclear matter.

The authors of[15] used the following model. Consider a potential with a Coulomb barrier (Fig. 2), within which a proton is in a quasi-stationary state with the proton decay energy Q_{odd} and the width γ. Now place another proton on the same level. Because of the proton-proton interaction U(1, 2) the total decay energy will not be $2Q_{odd}$, but $Q = 2Q_{odd} - E_{pair}$. It is clear from the foregoing discussion that for nuclei that are stable against the emission of a single (odd) proton but are unstable with respect to two-proton decay we have $1/2E_{pair} > B_{even} > 0$ (in other words, $2Q_{odd} > E_{pair} > Q_{odd}$). The proton pair is in a quasi-stationary state with the two-proton decay energy Q and width Γ. One proton alone cannot escape from this level, because even if it should leave the nucleus with zero energy it would not leave behind for its "partner" sufficient energy to occupy the only available level of energy Q_{odd}.

The complete Schrödinger equation is $H(1,2)\Psi(1,2) = E\Psi(1,2)$, where $H(1,2) = H_0(1) + H_0(2) + U(1,2)$, and $H_0(i)$ is the Hamiltonian of each proton ($i = 1, 2$). The two-proton function $\Psi(1,2)$ is decomposed into the single-proton radial functions $R_\epsilon(r)$:

$$\Psi(1,2)=\frac{1}{r_1r_2}\Omega(1,2)\int C(\varepsilon_1,\varepsilon_2)R_{\varepsilon_1}(r_1)R_{\varepsilon_2}(r_2)\,d\varepsilon_1\,d\varepsilon_2, \quad (6)$$

where $\Omega(1,2)$ is the angular part of the wave function. This leads to the customary equation in the theory of

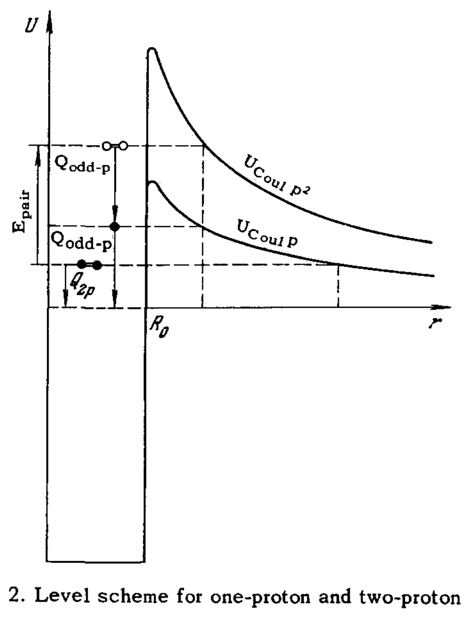

FIG. 2. Level scheme for one-proton and two-proton decays.

a superfluid nucleus:

$$(\varepsilon_1+\varepsilon_2)\,C(\varepsilon_1\varepsilon_2)-E_{\text{pair}}\sqrt{\Delta(\varepsilon_1)\Delta(\varepsilon_2)}\times\int C(\varepsilon_1',\varepsilon_2')\sqrt{\Delta(\varepsilon_1')\Delta(\varepsilon_2')}\,d\varepsilon_1'\,d\varepsilon_2'=EC(\varepsilon_1,\varepsilon_2), \quad (7)$$

where

$$\Delta\varepsilon=\frac{\gamma}{\pi[(\varepsilon-Q_{\text{odd}})^2+\gamma^2]},$$

and the constant E_{pair} is obtained from the proton interaction energy averaged over the angle variables $\overline{U}(1,2)$:

$$\langle\varepsilon_1,\varepsilon_2|\overline{U}(1,2)|\varepsilon_1',\varepsilon_2'\rangle\equiv-E_{\text{pair}}\sqrt{\Delta(\varepsilon_1)\Delta(\varepsilon_2)\Delta(\varepsilon_1')\Delta(\varepsilon_2')}. \quad (8)$$

Solving (7) in the customary manner, Galitskiĭ and Chel'tsov[15] obtained the total energy $E = Q - i\Gamma$ of the system, where

$$\Gamma=\frac{16}{\pi\sqrt{6}}Q\sqrt{\frac{\hbar v}{Ze^2}}\gamma^2\left(\frac{Q}{2}\right); \quad (9)$$

$\gamma(Q/2)$ is the width for one-proton decay with the energy $Q/2$ and ν is the proton velocity at this energy.

The exponential factor derived in[15] by this more exact procedure obviously agrees with our results in[9,10]. The pre-exponential factor, which was first calculated in[15], is

$$K_{2p}=\frac{2\Gamma}{\hbar}=\frac{8\hbar}{\pi\sqrt{6}}\frac{Q}{E^2_{\text{pair}}}\sqrt{\frac{\hbar v}{Ze^2}}K_p^2, \quad (10)$$

where $K_p = 2\gamma/\hbar \approx 10^{22}\ \text{sec}^{-1}$, so that for $Z = 20$ with Q and E_{pair} expressed in terms of MeV we have

$$K_{2p}=3.3\cdot10^{-22}K_p^2\frac{Q^{3/2}}{E^2_{\text{pair}}}\approx10^{22}\ \text{sec}^{-1}$$

with $Q \approx 1$ MeV and $E_{\text{pair}} \approx 2$ MeV.

As already mentioned, all the foregoing expressions were derived neglecting the possible role of a centrifugal barrier. It was shown in[16] that an essential change occurs when a centrifugal barrier is present, as, for example, in the case of the decay $Ge^{58} \rightarrow 2p + Zn^{56}$, when protons are emitted from the f shell. When the initial nucleus decays and the final nucleus is formed in its ground state the paired diproton is here emitted as an s wave, the centrifugal barrier being absent. When two protons are emitted separately, each having the same orbital angular momentum, the Coulomb barrier is accompanied by a centrifugal barrier.

Which has the greater probability—the emission of two protons, each having an energy Q/2 (so that the total energy is Q) and slowed down further by a centrifugal barrier, or the emission of a paired diproton subject only to a Coulomb barrier and having the energy $Q - \epsilon_0$? In the general case the exponential term of the decay constant is now

$$\exp\left\{-\frac{2}{\hbar}\left[\int_R^{r_0}\sqrt{2(2m)\left[\frac{2Ze^2}{r}-Q+\varepsilon_0\right]}\,dr+2\int_{r_0}^{R_{\text{max}}}\sqrt{2m\left[\frac{\hbar^2 l(l+1)}{2mr^2}+\frac{Ze^2}{r}-\frac{Q}{2}\right]}\,dr\right]\right\}, \quad (11)$$

where r_0 is the coordinate of effective diproton depairing under the potential barrier and R_{max} is the coordinate of the outside barrier edge for a given E. Introducing the notation

$$R_{\text{Coul}}=\frac{2Ze^2}{Q}\ \text{and}\ R_{\text{centr}}=\frac{\hbar}{\sqrt{mQ}}\sqrt{l(l+1)},$$

we obtain

$$R_{\text{max}}\approx R_{\text{Coul}}\left(1+\frac{R^2_{\text{centr}}}{R^2_{\text{Coul}}}\right),$$

which is an approximation that holds for $R^2_{\text{centr}} \ll R^2_{\text{Coul}}$ [i.e., for $Z^2 \gg 5l(l+1)Q$ MeV in practically all real cases].

It is easily shown that the minimum of the sum of the two integrals in (10), i.e., the most probable decay mode, is associated with

$$r_0=\frac{\hbar}{\sqrt{m\varepsilon_0}}\sqrt{l(l+1)}. \quad (12)$$

For $r_0 < R_{\text{max}}$ depairing of the diproton occurs under the barrier, at a distance greatly exceeding not only the nuclear radius R but also the amplitude of singlet nucleon-nucleon scattering or the effective size of a "free" diproton, $\hbar/\sqrt{m\epsilon_0} \approx 2.3 \times 10^{-12}$ cm. For $r_0 > R_{\text{max}}$ the proton pair proceeds as a unit through the barrier.

The pairing of the nuclear particles therefore leads to two effects in the presence of a centrifugal barrier: 1) enhanced barrier penetration compared with the passage of two independent particles, and 2) "restraining" by the barrier of the virtual singlet state of the nucleon pair at quite large distances (to almost 10^{-11} cm) with the consequent enhancing of the angular correlation of the particles.

The basic formulas for the penetration of mixed Coulomb and centrifugal barriers are given in[19] in accordance with the numerical data tabulated in[20]. We shall confine ourselves here to a single numerical example, the already mentioned hypothetical two-proton decay of Ge^{58}, for which $Q = 1.1$ MeV[10] and $l = 3$. In this case $R = R_p + R_{Ge} \approx 6.8 \times 10^{-13}$ cm, $R_{\text{Coul}} \approx 7.8 \times 10^{-12}$ cm, $R_{\text{centr}} \approx 2.1 \times 10^{-12}$ cm, $R_{\text{max}} \approx 8.4 \times 10^{-12}$ cm, and $r_0 \approx 8 \times 10^{-12}$ cm. Thus the depairing occurs under the potential barrier, close to its outside edge.

In the absence of a centrifugal barrier depairing would occur at the inside edge of the Coulomb barrier and the exponential decay-hindrance factor would be $\sim 10^{-22}$ in the present case. For the emission of two separate protons with $l = 3$ and the most probable energy $E_1 = E_2 = 0.55$ MeV the combined effect of the Coulomb and centrifugal barriers would here change the hindrance factor to 10^{-34}, thus making 2p decay entirely unobservable against the background of "superallowed" β^+ decay (the expected lifetime of Ge^{58} is ~ 0.1 sec). However, pairing leads to an extremely strong effect; the centrifugal barrier now reduces the decay rate by only one order of magnitude rather than by twelve orders as in the case of unpaired protons. The probability of 2p decay of Ge^{58} with the aforementioned value of Q is of the order 1%, i.e., the decay remains entirely observable. Therefore a centrifugal barrier should affect the 2p decay rate to an extremely smaller degree than the rate of "ordinary" one-proton decay.

The sharp rise, caused by pairing, of potential barrier penetration to two-particle tunneling should also lead to relatively large cross sections for the transfer of two neutrons or two protons in the nuclear reactions of heavy ions. The probability of nucleon-pair transfer with the "quenching" of the centrifugal barrier can be very close to the probability of one-proton transfer through a potential barrier that is heightened by orbital angular momentum. We have already compared two-proton radioactivity with electron tunneling from a superconductor to a normal metal. Continuing this analogy, the tunneling of a nucleon pair from one nucleus to another in reactions between heavy ions could be called the "nuclear Josephson effect."[21]*

3. ENERGY AND ANGULAR CORRELATION OF PROTONS IN TWO-PROTON DECAY

We have considered the energy correlation of protons in 2p decay,[9,10] in a review article,[11] and more thoroughly in[12,13]. This correlation should be the direct consequence of proton tunneling through a Coulomb barrier. As we have seen, its highest probability occurs when the decay energy is divided equally between the two protons.

When the proton energy is reduced by an amount δE the probability $W_b(E)$ of passage through the barrier penetration always increases by a greater amount than its increase accompanying the same enhancement (δE) of proton energy:

$$\frac{W_b(E)}{W_b(E-\delta E)} > \frac{W_b(E+\delta E)}{W_b(E)},$$

or $(W_b(E))^2 > W_b(E-\delta E)W_b(E+\delta E)$. We shall confine ourselves to the simplest and clearest case, which is the strongly sub-barrier case where not only $x \ll 1$, but also $\sqrt{x} \ll \pi/4$. Then in (5) we can neglect the term $2x^{1/2}$ in the square brackets and assume

$$C_{2p} = 2C_p = 2\pi\frac{Ze^2}{\hbar v} = 2\pi\frac{Ze^2\sqrt{m}}{\hbar\sqrt{Q}}.$$

In[12,13] we have considered the two-proton energy correlation for the general case where C_{2p} and C_p are represented by (1) and (2). However, *it is important* that the basic result obtained in the employed approximation and discussed below (the Gaussian distribution of proton energy around the most probable value Q/2), holds true even in the aforementioned general case.

Thus, for a pure Coulomb barrier, when diproton depairing takes place immediately at the nuclear boundary, the product of the two exponentials of barrier penetrability when the two protons have equal energies Q/2 is

$$W(0) = \exp\left\{-2\frac{2\pi Ze^2\sqrt{m}}{\hbar\sqrt{Q}}\right\} = W_{2p}, \qquad (13)$$

which is the same as the exponential for a diproton with the energy Q.

When one proton has the energy $Q(1+\kappa)/2$ and the other has $Q(1-\kappa)/2$, the product of the two exponentials is

$$W(\varkappa) = \exp\left\{-\frac{2\pi Ze^2\sqrt{m}}{\hbar\sqrt{Q}}\left(\frac{1}{\sqrt{1+\varkappa}}+\frac{1}{\sqrt{1-\varkappa}}\right)\right\}$$
$$\approx W(0)\exp\left\{-\frac{3\pi Ze^2\sqrt{m}}{2\hbar\sqrt{Q}}\varkappa^2\right\} = W(0)\,e^{-\alpha\varkappa^2} \qquad (14)$$

(the last relation results from the additional approximation $\kappa \ll 1$). We thus have a very simple Gaussian expression in which the coefficient $\alpha = 3\pi Ze^2\sqrt{m}/2\hbar\sqrt{Q}$ characterizes the degree of proton-energy correlation for the barrier. The half-width of the proton energy distribution is then

$$\Delta E = Q\sqrt{\frac{\ln 2}{\alpha}} = 2Q\sqrt{\ln 2}\left(\frac{\hbar\sqrt{Q}}{6\pi Ze^2\sqrt{m}}\right)^{1/2}. \qquad (15)$$

Galitskiĭ and Chel'tsov[15] also obtained the proton energy distribution in 2p decay for a pure Coulomb barrier by solving the nonstationary Schrödinger equation. The exact solution for the distribution function $dW/d\kappa$ is[15]

$$\frac{dW}{d\varkappa} = \frac{4\hbar}{\pi}\frac{QE_{\text{pair}}^2 \quad K_p^2}{[E_{\text{pair}}^2 \quad -\varkappa^2Q^2]^2}W(0)\exp\left\{-\frac{3\pi Ze^2\sqrt{m}}{2\hbar\sqrt{Q}}\varkappa^2\right\}, \qquad (16)$$

which agrees with the approximations in[9-13] for the magnitude of the exponential factor.

It should be noted that the form of the proton-energy correlation should be extremely sensitive to the form of the Coulomb barrier. A diffuse edge of the potential well, or the possibility of energy exchange between the two protons when one of them is inside and the other outside the nucleus in the case of a Coulomb barrier (represented by the "tail" of the wave function), leads to enhanced barrier penetration, i.e., to a reduction of the correlation coefficient α from its maximum value, which in the general case is

$$\alpha_{\max} = \frac{Ze^2\sqrt{m}}{\hbar\sqrt{Q}}\left[3\arccos x^{1/2} + \frac{(3-x)\,x^{1/2}}{(1-x)^{1/2}}\right]. \qquad (17)$$

The character of the correlation also varies greatly when the Coulomb barrier is accompanied by a centrifugal barrier affecting separate protons but not affecting a proton pair. In this case the two protons traverse a considerable fraction or their entire sub-barrier path in the paired form, and the diproton breaks apart,

*The Josephson effect is the tunneling of Cooper pairs from one superconductor to another through a potential barrier. This tunneling of pairs is characterized by high probability that is fully commensurable with the probability of tunneling by single electrons.

yielding the energy $\epsilon_0 \approx 70$ keV of the nucleon-nucleon virtual $^1S_0$ level, at a distance $r_0 \approx 2.3\sqrt{l(l+1)} \times 10^{-12}$ cm from the center of the nucleus or at the outside edge of the barrier. The energy spread of the two protons is then given by the difference between the two quantities

$$E_{\max}=\left(\sqrt{\frac{Q}{2}}+\sqrt{\frac{\varepsilon_0}{2}}\right)^2 \text{and}\, E_{\min}=\left(\sqrt{\frac{Q}{2}}-\sqrt{\frac{\varepsilon_0}{2}}\right)^2,$$

so that the half-width of the energy distribution is

$$\Delta E=\frac{1}{2}(E_{\max}-E_{\min})=\sqrt{\varepsilon_0 Q}\,. \qquad (18)$$

It is interesting that in some instances the addition of a centrifugal barrier to the Coulomb barrier weakens the energy correlation; thus, in the foregoing case of Ge^{58} we obtain $\Delta E \approx 0.27$ MeV instead of $\Delta E \approx 0.17$ MeV for a pure Coulomb barrier. Then the relative width of the energy distribution, $\Delta E/(Q/2)$, will decrease as $Q^{-1/2}$ with increasing 2p decay energy for a mixed barrier, but will increase as $Q^{1/4}$ for a pure Coulomb barrier.

The character of the angular correlation between the two protons emitted in 2p decay was investigated in[12,13] using a method developed by A. B. Migdal[22] for nuclear reactions accompanied by the formation of a pair of slow nucleons. If we begin by neglecting the Coulomb interaction between the emitted protons, our problem becomes similar to the formation of neutron pairs that was considered by Migdal. In this case we have

$$\frac{df(\vartheta)}{d\Omega}=\Phi(\vartheta)\approx\frac{1-\operatorname{erf}\sqrt{a(\varepsilon_0+\gamma)}}{\sqrt{\varepsilon_0+\gamma}}\exp\left\{\frac{1}{2}\alpha\vartheta^2\frac{\varepsilon_0}{Q}\right\}, \qquad (19)$$

where ϑ is the lab-system angle between the two emitted proton directions, $d\Omega = 2\pi \sin\vartheta\, d\vartheta$, $\gamma = Q\vartheta^2/4$, and $a = (4/3)\alpha/(Q+\gamma)^{3/2}$; α is given through (14). The difference from Migdal's formulas for two neutrons, caused by taking into account the passage of the emitted protons through the Coulomb barrier of a radioactive nucleus, ordinarily reduces to small corrections; the exponential factor is also close to unity because, for example, when $Z = 20$ and $Q = 1$ MeV we have $\alpha \approx 20$ and $\frac{1}{2}\alpha\, \epsilon_0/Q \approx 0.7$. Consequently, the distribution at small angles ϑ in the considered approximation is close to that obtained for two neutrons in[22]:

$$\Phi(\vartheta)\propto\frac{1}{\sqrt{\frac{\varepsilon_0}{Q}+\left(\frac{\vartheta}{2}\right)^2}}=\frac{\sqrt{Q}}{\sqrt{\varepsilon_0+\gamma}}\,, \qquad 20)$$

and the half-width of the distribution corresponds to $\Delta\vartheta = 2\sqrt{3\epsilon_0/Q}$ or $\Delta\vartheta \approx 1/\sqrt{Q(\mathrm{MeV})}$.

The Coulomb interaction between the emitted protons broadens the angular distribution appreciably; the distribution function $\Phi(\vartheta)$ here depends on γ as follows:[22]

$$\gamma(\mathrm{MeV})=0 \quad 1 \quad 2 \quad 3 \quad 4 \quad 5,$$
$$\Phi(\vartheta)=1 \quad 0.82 \quad 0.59 \quad 0.40 \quad 0.34 \quad 0.31,$$

so that we have for the half-width $\Delta\vartheta \approx 2\sqrt{3/Q(\mathrm{MeV})}$.

The presence of a centrifugal barrier should, as a rule, strengthen the angular correlation of protons in 2p decay. If depairing occurs only at the outside barrier edge $R_{\max}$ (for $r_0 > R_{\max}$) we have the angular distribution half-width $\Delta\vartheta \approx \sqrt{\epsilon_0/Q}$.

In the case of Ge^{58} decay ($Q \approx 1.1$ MeV), which has already been cited as an illustration, the centrifugal barrier practically converts the isotropic angular distribution into the characteristic pattern of two protons emitted in almost the same direction forming a mean angle $\Delta\vartheta \approx 0.25$.

4. POSSIBLE TWO-PROTON RADIOACTIVE NUCLEI

Before proceeding to enumerate the possible 2p-radioactive nuclei it will be useful to consider the different decay modes, which are represented schematically in Fig. 3.

The simplest mode (I) is $Q_{odd} > 0$, $Q_{even} < 0$, $Q_{2p} > 0$, where the initial even-Z nucleus is completely

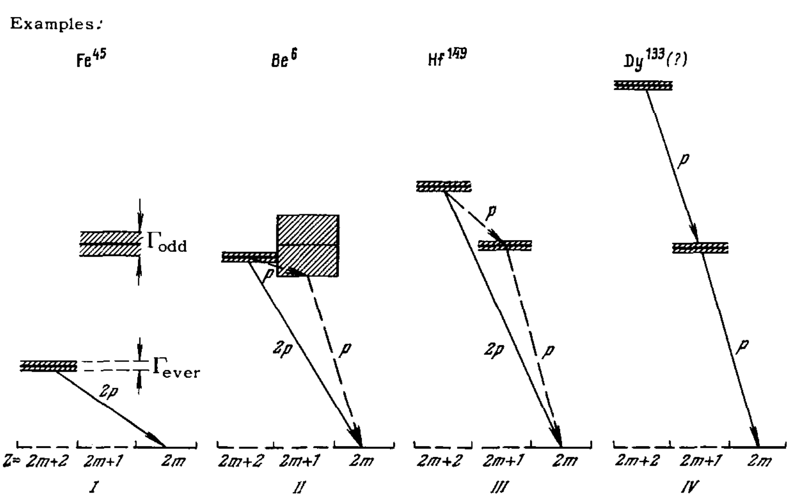

FIG. 3. Different modes of proton pair emission.

stable against one-proton emission:

$$Q+\frac{1}{2}\Gamma_{\text{even}}<Q_{\text{odd}}-\frac{1}{2}\Gamma_{\text{odd}} \quad \text{or} \quad Q<Q_{\text{odd}}-\frac{1}{2}\Gamma,$$

where $\Gamma = \Gamma_{even} + \Gamma_{odd}$, and Γ_{even} and Γ_{odd} are the respective widths of the initial even-Z nucleus and the odd-Z daughter nucleus. In this case we obviously have the inequalities

$$E_{\text{pair}}>Q_{\text{odd}}>\frac{1}{2}E_{\text{pair}}>B_{\text{even}}>0 \text{ and } E_{\text{pair}}>Q>0.$$

We now fix the decay energy Q_{odd} of the odd daughter nucleus and elevate the energy Q of two-proton decay.

The next mode, that of Be^6, is subject to the additional condition $B_{even} < \Gamma/2$ (where $\Gamma < E_{pair}$ is assumed) and to the inequalities

$$E_{\text{pair}}>Q_{\text{odd}}>E_{\text{pair}}-\frac{\Gamma}{2},\ E_{\text{pair}}>Q_{2p}>Q_{\text{odd}}-\frac{\Gamma}{2}.$$

Here, despite the energy instability of the initial nucleus with respect to one-proton decay, the overlapping of levels permits both two-proton decay and the successive emission of two protons. To estimate the decay rate we can take as the effective one-proton decay energy of the initial even nucleus

$$Q_{\text{even eff.}}=\frac{\Gamma}{2}-B_{\text{even}}.$$

This decay mode has the highest probability for the lightest nuclei possessing 2p instability, because the corresponding daughter nuclei are characterized by the greatest widths for the emission of an "odd" proton.

The third and last mode (III) of two-proton decay is characterized by the conditions

$$Q_{\text{odd}}>0, \quad Q_{\text{even}}>0, \quad Q_{2p}>0, \qquad \text{(20f)}$$

i.e., by energetic instability of the initial nucleus with respect to not only two-proton decay, but also proton decay followed by proton emission from the odd daughter nuclide.

However, since the energy of the second decay event $Q_{odd} = Q_{even} + E_{pair}$ greatly exceeds the energy Q_{even} of the primary transformation, one-proton emission from an even-Z nucleus followed by the emission of a second proton will often be exponentially smaller than direct two-proton decay with $Q = 2Q_{even} + E_{pair}$. Therefore this mode of 2p radioactivity, which is especially characteristic of heavier nuclei and is the only mode possible for Z > 50,[17] is associated with values of Q that exceed the proton pairing energy.

For sufficiently large values of Q_{odd}, on the other hand, the dominant mode is the successive emission of two protons (mode IV in Fig. 3). It is interesting that this mode, which seems at first glance to be the most natural and most widely occurring mode, becomes a very rare exception as a result of the requirement $\tau > 10^{-12}$ sec. For even Z > 50, α decay is energetically possible in addition to two-proton decay or two successive proton decays. For odd Z > 50 this decay mode competes only with one-proton decay (neglecting the slower β^+ decay). However, as has recently been shown in[23], p decay should dominate in many instances.

In order to determine which one of the three enumerated decay modes involving the emission of heavy charged particles is the principal mode for even nuclides, we must make use of Fig. 4 (see the caption).

It must also be remembered that the presence of both the centrifugal and Coulomb barriers suppresses mainly one-proton decay and should thus favor α and 2p decays. An illustration is provided by the curves in Fig. 4 for Z = 60 and l = 4 (proton emission from the g shell).

How can two-proton radioactive decay be distinguished from two successive p-decay events in future experiments? The conventional method of delayed coincidences is here often ineffectual because of the extreme rapidity with which the emission of the second proton follows the first (to $\tau \approx 10^{-19}$—10^{-20} sec). However, the energy and angular characteristics of the emitted protons are completely reliable criteria. Instead of two proton lines with the energies Q_{even} and Q_{odd} belonging to successive p decays the most probable proton energy in 2p decay will be $Q/2 = 1/2 \times (Q_{even} + Q_{odd})$; the previously discussed energy and

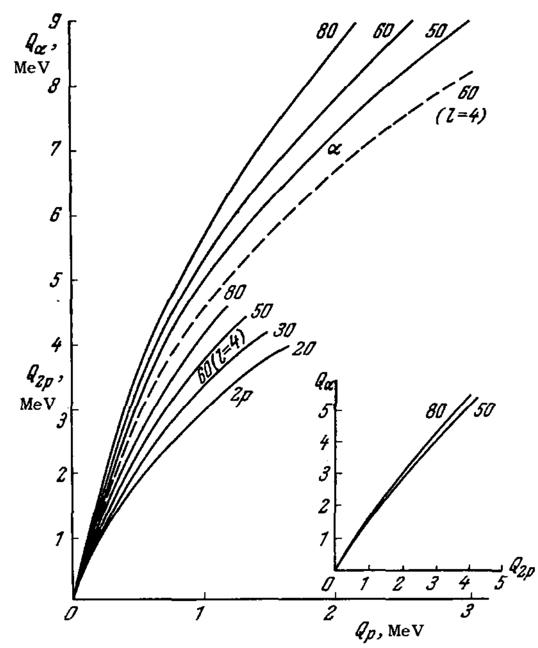

FIG. 4. Relations between the energies Q_α of α decay, Q_{2p} of two-proton decay, and Q_p of proton decay for different values of Z (shown at the end of each curve), corresponding to equality of the three decay rates for a pure Coulomb barrier, and for the special case l = 4 (Z = 60) when a centrifugal barrier is also present for single protons. In the larger figure the upper curves relate Q_α and Q_p, while the lower curves relate Q_{2p} and Q_p. The smaller graph at the right gives the relation between Q_α and Q_{2p}.

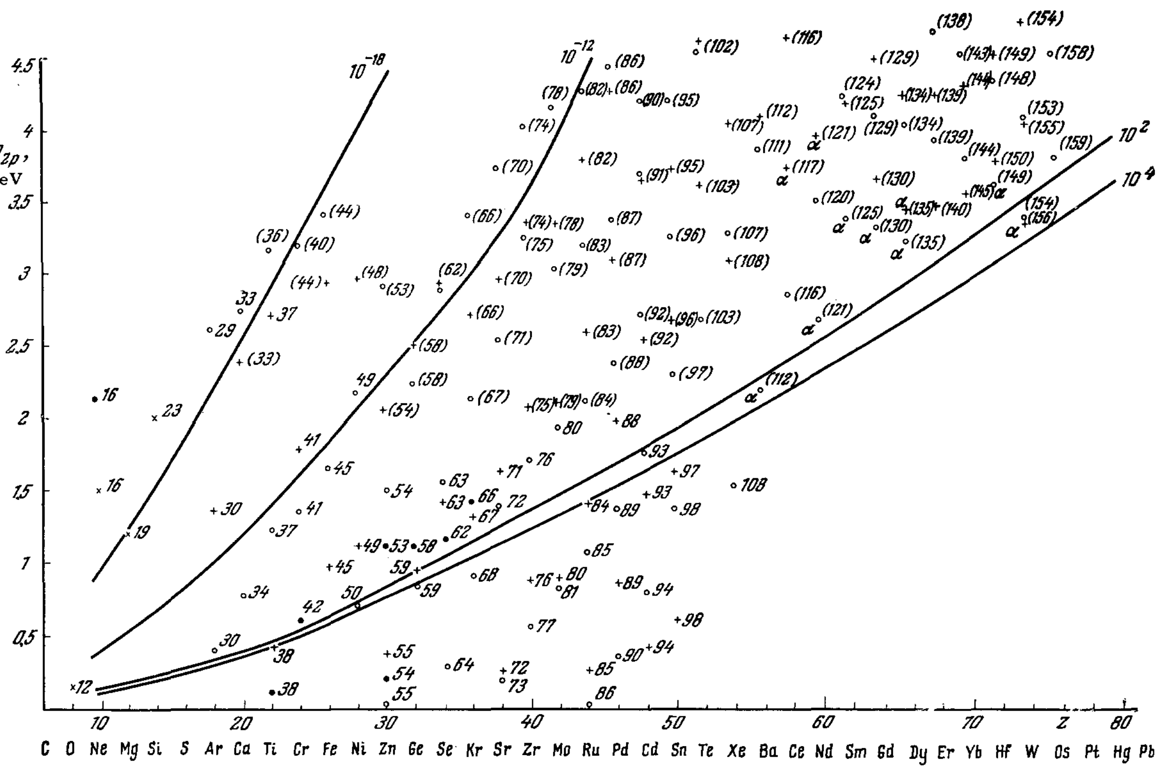

FIG. 5. Two-proton decay energies of neutron-deficient isotopes of even elements from oxygen to osmium (Z = 8–76). The sources of the energy values are indicated as follows: • – [9,10], × – [14], + – [24], and ∘ – [25]. The four calculated curves correspond to the partial lifetimes $\tau_{2p} = 10^{-18}$, 10^{-12}, 10^{2}, and 10^{4} sec for a pure Coulomb barrier. The mass numbers of the nuclei that are unstable with respect to one-proton decay are given in parentheses. The symbol α denotes possible strong competition with α decay.

angular correlations of the two protons should also be observed. We now turn to a specific enumeration of the possible 2p-radioactive nuclei. We take as the lower lifetime limit for determining radioactivity $\tau = 10^{-12}$ sec. The upper limit is given by the competition with β^+ decay. For all neutron-deficient nuclei with Z > N superallowed β^+ decay is possible, limiting the overall half-lives to ~ 0.1—0.01 sec (decreasing as Z increases). Therefore, as the upper limit of the observed lifetime with respect to 2p decay for Z > N we can take $\tau \approx 10^2$ sec. When superallowed β^+ decay is impossible (for Z < N) this limit can be raised to $\approx 10^4$ sec.

For all two-proton radioactive nuclei up to tin we have Z > N and superallowed β^+ decay is possible. Therefore we cannot hope here for the existence of 2p radioactive nuclei with lifetimes exceeding tens or hundreds of milliseconds.

In the region of elements heavier than tin superallowed β^+ decay should not occur in 2p-radioactive nuclei with $Z \leq N$, and lifetimes of tens or hundreds of seconds are possible. In accordance with the foregoing, Fig. 5, which summarizes the possible two-proton radioactive nuclei, shows curves corresponding to 10^4, 10^2, 10^{-12}, and even 10^{-18} sec partial lifetimes with respect to 2p decay (for a pure Coulomb barrier, i.e., for the effective 2p-decay energy Q rather than $Q - \epsilon_0$ and for 10^{-22} sec as the factor before the exponential). The two-proton decay energies were taken from our own work in[9,10], Jänecke's article,[14] and the mass tables of Cameron[24] and Seeger.[25] The calculations seem to be most accurate when the method proposed in[9,10] can be used to determine the proton binding energy in a neutron-deficient nucleus.

A direct consequence of isotopic invariance is the following simple formula[9-11] relating the binding energy B_p of the Z-th proton in a nucleus ${}_ZM_N^A$ and the binding energy B_n of the Z-th neutron in the mirror nucleus ${}_NM_Z^A$ to the difference ΔB_0 between the neutron and proton binding energies in the isotopically self-conjugate nucleus containing Z neutrons and Z protons:

$$\Delta B_{np} = B_n({}_NM_Z^A) - B_p({}_ZM_N^A)$$

$$= B_n({}_ZM_Z^{2Z}) - B_p({}_ZM_Z^{2Z}) = \Delta B_0. \quad (21)$$

Thus the difference ΔB_{np} is determined completely by Z, being almost entirely independent of N:

$$\Delta B_{np} \approx 1.2 \frac{Z-1}{(2Z-1)^{1/3}} \left\{ 1 + \left(\frac{A-2Z}{3A} \right)^2 \left(1 + \frac{1}{A-2Z} \right) + \ldots \right\}$$

$$\approx 1.2 \frac{Z-1}{(2Z-1)^{1/3}} \text{ MeV}. \quad (22)$$

We shall now illustrate the accuracy of the predictions based on Eq. (21) with only two examples. The tables in[26] give a mass defect 8.28 MeV (on the C^{12} scale) for Na^{20}. The application of (21) to data already known regarding the nucleon binding energies in F^{20}, Ne^{21}, and O^{19} (as discussed more thoroughly in[8]) indicates that this value is too high by 1.4—1.5 MeV and that the mass defect of Na^{20} is 6.78—6.88 MeV. A recent experiment yielded 6.83 ± 0.06 MeV.[27]

The neutron binding energy in Li^9 is 4.054 MeV[28] and ΔB_0 (C^{12}) = 2.765 MeV. We therefore obtain B_p (C^9) = 1.29 MeV, and 28.92 MeV for the mass defect of C^9 compared with the experimental value 28.95 ± 0.15 MeV.[29]

On the basis of (21) and certain data for light neutron-rich nuclei we can examine more thoroughly the isotopes O^{12}, Ne^{16}, and Mg^{19}, which may be 2p-unstable according to Zel'dovich.[5] Jänecke[14] gives the 2p-decay energy of O^{12} as $Q \approx 0.15$ MeV, which would correspond to 2p radioactivity and not simply to instability (Fig. 5). However, let us consider Be^{12}. From considerations given in[30] based on the systematics of neutron pairing energy this energy should lie between the limits 1.5 MeV (E_{pair} for B^{13}) and 5.15 MeV (for B^{10}), i.e., B_n (Be^{12}) = 2—5.65 MeV. Since ΔB_0 (O^{16}) = 3.54 MeV, we obtain B_p (O^{12}) $\approx$ (−1.5)—2.1 MeV. Similarly, knowing B_n (Be^{11}) = −0.5 MeV and ΔB_0 (N^{14}) = 3 MeV, we obtain B_p (N^{11}) = −2.5 MeV. Therefore the 2p-decay energy of O^{12} clearly exceeds 0.4 MeV. Using the experimental[31] β^- decay energy of B^{12} (11.7 MeV), we obtain Q = 2.9 MeV for the 2p-decay energy of O^{12}. Since the ground state of N^{11} is very broad, the case of O^{12} is similar to that of Be^6.

A similar but more definite conclusion is reached for Ne^{16}. In its mirror nucleus C^{16} we have the neutron binding energy B_n = 4.25 MeV,[31] so that B_p (Ne^{16}) = 0.2 MeV. The F^{16} nucleus is highly unstable and must be characterized by a very broad ground level: Q_{odd} = 2.3 MeV. Finally, Q_{2p} (Ne^{16}) $\approx$ 2.1 MeV, which corresponds to a lifetime less than 10^{-18} sec.

We must here mention recent experimental work[18] on the possibility of Ne^{16} production through the transfer of four neutrons when nickel is bombarded with 150-MeV Ne^{20} ions.

The hypothetical Ne^{16} ions were collimated by the magnetic field of a cyclotron and struck an emulsion which could have registered the successive emission of two protons. Not a single instance of such emission was observed. The authors of[18] therefore concluded that the lifetime of Ne^{16} is shorter than 10^{-8} sec, or that if its lifetime is longer the cross section for Ne^{16} production is smaller than 1.8×10^{-30} cm^2.

Mg^{19} is proton-stable if B_n (N^{19}) > 4.8 MeV and is two-proton stable if B_n (N^{19}) > 6.3 MeV because Q_{odd} (Na^{18}) $\approx$ 1.5 MeV.

By extrapolating the binding energy of the 12th neutron in the series

	Mg^{24}	Na^{23}	Ne^{22}	F^{21}	O^{20}	N^{19}
B_n(MeV) =	16.6	12.4	10.4	8.1	7.6	?

we are led to the conclusion only that Mg^{19} is p-stable, while leaving open the question of its 2p instability or 2p radioactivity. Since the neutron pairing energy in N^{19} should be smaller than in O^{20} (3.6 MeV) and N^{17} (3.37 MeV) and for the neutron binding energy in N^{18} we have B_n (N^{18}) = 2.84 ± 0.4 MeV,[32] it follows that B_n (N^{19}) < 6.2 ± 0.4 MeV. Therefore Mg^{19} appears to be actually 2p unstable and may even be 2p radioactive.

Si^{23}, the next isotope shown in Fig. 5, is highly 2p unstable according to[14]. Using (21), we easily obtain the binding energies of proton pairs in the heavier silicon isotopes:

	Si^{23}	Si^{24}	Si^{25}	Si^{26}
B_{2p}(MeV) =		3.4	5.5	7.7.

The extrapolation of the values of B_{2p} to Si^{23} raises doubts about the 2p instability of this isotope, especially because of the large value Q = 2 MeV that is shown in Fig. 5.

We shall not discuss other examples in the same detail, but note only that the data in the generally useful tables of Cameron and of Seeger are sometimes clearly incorrect. For example, the mass defects of Ga^{37}, Ga^{38}, and Ti^{41} are too small by more than 2 MeV, and the mass defect of As^{66} is too high by about the same amount. The corresponding predictions in Fig. 5 are therefore only rough approximations.

We note, furthermore, that the total number of 2p-radioactive nuclei may be greater than 60, of which about half are in the region Z > 50, with 2p-decay energies exceeding the pairing energy and with lifetimes that have no upper limit imposed by superallowed β^+ decay.

The most practical method of producing 2p-radioactive nuclei makes use of reactions involving multiply-charged heavy ions, such as Ca^{40} (Ca^{40}, 4n), Zr^{76} or Ni^{58} (Ni^{58}, 4n), Ba^{112}. In some instances it may be convenient to use (He^3, xn) or (p, yn) reactions.

The utilization of heavy ions has already led to the discovery of delayed proton emission and to the successful investigation of this effect. We therefore expect that the discovery and study of two-proton radioactivity will take place in the not too distant future.

In addition to the investigation of the levels and sizes of neutron-deficient nuclei accompanying the study of proton radioactivity (which are important for the collective model of the nucleus), the investigation of two-proton radioactivity provides very definite possibilities for studying the shape of the potential barrier around nuclei, proton pairing below the barrier, and new nuclear phenomena related to those observed in the study of superconductivity. For these reasons two-proton radioactivity has aroused wide interest and will be searched for by laboratories in different countries.

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